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(54) Production process for glycidyl ether adduct and catalyst used for the process

(57) Provided is a catalyst for adding a glycidyl ether to an active hydrogen-containing organic compound, comprising a complex oxide of magnesium and at least one element other than magnesium selected from the group consisting of the elements in the third period and the fourth period in the periodic table. Also provided is a process for producing a glycidyl ether adduct, which comprises subjecting an active hydrogen-containing organic compound and a glycidyl ether to an addition reaction in the presence of the above-mentioned catalyst.

In the addition reaction between the active hydrogen-containing organic compound and the glycidyl

ether, use of the catalyst of the present invention enables inhibition of an excess addition reaction of the glycidyl ether which is a successive reaction, and a mono- or di-(alkyl, alkenyl or phenyl) ether product in which one or two glycidyl ethers are added is selectively produced by adjusting the mole ratio of the active hydrogen-containing organic compound to the glycidyl ether to a specific range. Accordingly, the mono- or di-(alkyl, alkenyl or phenyl) ether product can be obtained at a high productivity with a high purity.

Description**Field of the Invention**

5 [0001] The present invention relates to a process for producing a glycidyl ether adduct which is useful as a surfactant to be incorporated in formulations such as detergents and cosmetics, and to a catalyst used for such process.

Background of the Invention

10 [0002] In general, an addition reaction between a glycidyl ether and an organic compound having active hydrogen atoms is carried out in the presence of a basic catalyst. Active hydrogen atoms originating in a glycidyl ether are newly produced in the reaction product, and therefore a problem is created in which the excess glycidyl ether is further reacted with the newly produced reaction product having active hydrogen atoms. For example, a reaction between glycerin and a glycidyl ether is described in Japanese Patent Application Laid-Open (through PCT) No. 500861/1995, and in 15 this case, glycerin is used in an amount of five times the moles based on the amount of the glycidyl ether in order to inhibit the excess addition reaction described above. However, excess use of glycerin results in, as a matter of course, a reduction in the productivity and an increase in the cost required for recovery, and therefore is not preferred from an industrial viewpoint.

Summary of the Invention

20 [0003] The present invention provides a catalyst for an addition reaction between an active hydrogen-containing organic compound and a glycidyl ether, which comprises a complex oxide of magnesium and at least one element other than magnesium selected from the group consisting of the elements in the third period and the fourth period in 25 the periodic table.

[0004] Further, the present invention provides a process for producing a glycidyl ether adduct, which comprises subjecting an active hydrogen-containing organic compound and a glycidyl ether to an addition reaction in the presence of a catalyst comprising a complex oxide of magnesium and at least one element other than magnesium selected from the group consisting of the elements in the third period and the fourth period in the periodic table.

Detailed Description of the Invention

30 [0005] The present inventors have found that, by using a specific complex metal oxide as a catalyst, it is possible to efficiently provide a desired glycidyl ether adduct through an addition reaction between a glycidyl ether and an organic compound having active hydrogen atoms even if the organic compound is used in a small excess amount. Further, the present inventors have found that a product in which one glycidyl ether is added or a product in which two glycidyl ethers are added can be synthesized by changing the mole ratio of the organic compound to the glycidyl ether.

35 [0006] In the catalyst of the present invention, the elements in the third period and the fourth period in the periodic table excluding magnesium include Na, Al, Si, K, Cu, Ca, Zn, Ga, Sc, Ti, V, Ge, Se, Cr, Mn, Fe, Co and Ni. Among 40 them, Al, Zn, Cr, Fe, Co and Ni are preferred, and Al and Zn are more preferred.

[0007] The complex oxide of Mg (magnesium) and at least one of the elements described above is preferably an oxide which comprises Mg as a principal component and to which at least one of the elements described above excluding Mg is added.

45 [0008] A blending proportion of the magnesium to the element(s) other than magnesium in the third period and/or the fourth period in the periodic table which are contained in the catalyst of the present invention is such a proportion that the amount of the element(s) other than magnesium in the third period and/or the fourth period in the periodic table is preferably from about 0.005 to 0.4 atom, particularly preferably from about 0.01 to 0.3 atom per atom of magnesium which is a principal component. A trace amount of a third component may be added to the catalyst as long as such addition does not adversely affect the catalyst of the present invention.

50 [0009] The catalyst of the present invention may consist essentially of the complex oxide described above or may comprise a carrier on which the complex oxide is supported. The carrier usable in the invention includes diatomaceous earth, zeolite, mordenite, montmorillonite, tin oxide, titanium oxide and activated carbon. The amount of the complex oxide supported on the carrier is preferably from about 10 to 80 % by weight, more preferably from about 20 to 60 % by weight of the carrier.

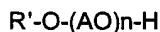
55 [0010] A process for producing the catalyst of the present invention is not particularly restricted, and it is produced by a publicly known process. For example, it is produced by a co-precipitation process in which a precipitating agent is added to a mixed aqueous solution or an aqueous dispersion containing compounds such as chlorides, hydroxides, oxides, nitrates, sulfates or carbonates of the respective elements, or a precipitating agent is added to a mixed aqueous

5 solution or an aqueous dispersion containing compounds capable of changing to catalyst components other than a carrier component in the presence of a fine powdery carrier to obtain a precipitate and then the precipitate is washed with water, dried and calcined. Alternatively, the catalyst is produced by an impregnation process in which a fine powdery carrier is impregnated with an aqueous solution or an aqueous dispersion of compounds capable of changing to catalyst components other than a carrier component and the resulting carrier is then dried and calcined. When the catalyst is produced by the co-precipitation process or the impregnation process, any compounds can be used for the above-mentioned compounds as long as they are water-soluble or water-dispersible.

10 [0011] A specific example of the co-precipitation process used for preparing the catalyst of the present invention includes, for example, a process in which, to a mixed aqueous solution or aqueous dispersion containing a magnesium compound selected from chloride, hydroxide, oxide, nitrate, sulfate and carbonate of magnesium and a compound selected from chlorides, hydroxides, oxides, nitrates, sulfates and carbonates of at least one element other than magnesium selected from the elements in the third period and the fourth period in the periodic table, preferably a mixed aqueous solution or aqueous dispersion containing magnesium nitrate, magnesium chloride, magnesium sulfate or magnesium hydroxide and carbonates, hydroxides or oxides of at least one element other than magnesium selected from the elements in the third period and the fourth period in the periodic table, more preferably a mixed aqueous solution or aqueous dispersion containing magnesium nitrate, magnesium chloride, magnesium sulfate or magnesium hydroxide and carbonate, hydroxide or oxide of aluminum or zinc, an aqueous solution of an alkaline agent selected from hydroxides or carbonates of ammonia or alkaline metals, preferably hydroxides of alkaline metals, is added in an amount of from about 0.8 to 1.2 times equivalent based on the amount of the compound(s) of the element(s) other than magnesium selected from the elements in the third period and the fourth period in the periodic table to adjust the pH to 6 to 11, preferably 7 to 10, to give hydroxides, carbonates or oxides of the element(s) or a mixture thereof by a precipitation reaction, and after removing water soluble salts by washing with water, the hydroxides, carbonates or oxides of the element(s) or the mixture thereof are calcined at a temperature in a range of from about 400 to 900°C, preferably from about 450 to 700°C. Further, a specific example of the impregnation process used for preparing the catalyst of the present invention includes a process in which a carrier, carbonate, hydroxide or oxide of magnesium and a carbonate, hydroxide or oxide of at least one element other than magnesium selected from the elements in the third period and the fourth period in the periodic table are mixed in an aqueous medium, dried and then calcined at a temperature in a range of from about 400 to 900°C, preferably from about 450 to 700°C. These oxides may be separately prepared and mixed in a required proportion for use. Among these preparation processes, the co-precipitation process is particularly preferred.

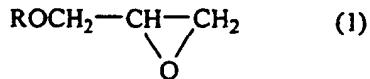
15 [0012] The active hydrogen-containing organic compound used for the process for producing a glycidyl ether adduct according to the present invention is not particularly restricted as long as it contains active hydrogen atoms and produces a glycidyl ether adduct by the reaction with a glycidyl ether. Specifically, it includes hydroxyl compounds, thiols, carboxylic acids, amines, amides and mixtures thereof.

20 [0013] Among them, the hydroxyl compounds are preferred. The hydroxyl compounds include preferably linear or branched monohydric alcohols having 1 to 30 carbon atoms, polyols having 2 to 18 carbon atoms and 2 to 18 hydroxyl groups and ketals thereof, and polyoxyalkylene alkyl ethers, more preferably polyols having 2 to 12 carbon atoms and 2 to 12 hydroxyl groups and ketals thereof, and still more preferably ethylene glycol, propanediol, glycerin, pentaerythritol, diglycerin, polyglycerin, sorbitol, glucose, sucrose and glycerin ketal. Mixtures thereof are also preferred. The polyoxyalkylene alkyl ethers have preferably the following formula:



25 [0014] wherein R' represents a linear or branched alkyl group having 1 to 30 carbon atoms; A represents an alkylene group having 2 or 3 carbon atoms; n represents an integer of from 1 to 50, and plural A's may be the same or different. In the formula described above, preferably, R' represents a linear or branched alkyl group having 2 to 18, particularly 2 to 12 carbon atoms; A represents an alkylene group having 2 carbon atoms; and n represents an integer of from 1 to 20, particularly 1 to 10.

30 [0014] The glycidyl ether used in the present invention includes a compound represented by Formula (1):



wherein R represents a linear or branched alkyl or alkenyl group having 1 to 36 carbon atoms or a phenyl group. In

Formula (1), R is an alkyl or alkenyl group having preferably 1 to 20, more preferably 4 to 18 carbon atoms, and is particularly preferably an alkyl group having 4 to 18 carbon atoms. Specific examples of preferred R include butyl, pentyl, 2-methylbutyl, hexyl, octyl, 2-ethylhexyl, decyl, dodecyl, hexadecyl and methylheptadecyl (isostearyl).

[0015] The glycidyl ether adduct obtained in the present invention is preferably a monoalkyl (or alkenyl or phenyl) ether product in which one glycidyl ether is added or a dialkyl (or alkenyl or phenyl) product in which two glycidyl ethers are added.

[0016] The addition reaction of the glycidyl ether using the catalyst of the present invention can readily be carried out according to a conventional operating procedure under conventional reaction conditions. The reaction temperature is preferably from about 80 to 230°C, more preferably from about 120 to 200°C. The amount of the catalyst varies depending on the mole ratio of the starting materials and is usually such an amount that the complex oxide described above amounts to preferably from about 0.05 to 20 % by weight, more preferably from about 1 to 10 % by weight based on the amount of the glycidyl ether.

[0017] When producing the monoalkyl (or alkenyl or phenyl) ether product in which one glycidyl ether is added, a feeding proportion of the glycidyl ether to the organic compound having active hydrogen atoms falls preferably in a range of from about 1 : 0.9 to 1 : 3, particularly from about 1 : 0.9 to 1 : 1.5 in terms of a mole ratio from the viewpoint of a rise in the productivity. On the other hand, when producing the dialkyl (or alkenyl or phenyl) product in which two glycidyl ethers are added, a feeding proportion of the glycidyl ether to the organic compound having active hydrogen atoms falls preferably in a range of from about 1.8 : 1 to 3 : 1, particularly from about 1.8 : 1 to 2.5 : 1 in terms of a mole ratio from the viewpoint of a rise in the productivity.

[0018] The reaction is preferably carried out under an atmosphere of an inert gas such as, for example, nitrogen gas. The reaction pressure is preferably from about 0.1 to 20 atm, more preferably from about 0.9 to 10 atm.

[0019] In the reaction operation, for example, a reactor is charged with the raw material of the active hydrogen-containing organic compound and the catalyst, and after substituting with nitrogen, a glycidyl ether is introduced thereto at a predetermined temperature to effect a reaction between them. A method for separating the catalyst is not specifically restricted, and considered is a method in which the reaction product obtained after the reaction is cooled and water for reducing the viscosity and a filtering aid (diatomaceous earth, a cellulose aid, activated clay or the like) are added to filter the catalyst. The preferred product obtained by the process of the present invention is substantially neutral and therefore need not be neutralized by adding an acid or an alkali.

[0020] The following examples further describe and demonstrate embodiments of the present invention. The examples are given solely for the purpose of illustration and are not to be construed as limitations of the present invention.

Examples

[Catalyst preparation]

Catalyst 1

[0021] A complex oxide having a chemical composition of $Mg_6Al_2(OH)_{16}CO_3 \cdot 4H_2O$ (Kyoward 500 manufactured by Kyowa Chemical Co., Ltd.) (15g) was activated by calcining at 450°C for 2 hours to obtain 8.9g of a catalyst powder.

Catalyst 2

[0022] A complex oxide having a chemical composition of $Mg_{0.7}Al_{0.3}O_{1.15}$ (Kyoward 2000 manufactured by Kyowa Chemical Co., Ltd.) was used as a catalyst as it is.

Catalyst 3

[0023] Aluminum hydroxide-magnesium having a chemical composition of $5MgO \cdot Al_2O_3 \cdot mH_2O$ (Kyoward 300 manufactured by Kyowa Chemical Co., Ltd.) (15g) was activated by calcining at 550°C for 2 hours to obtain 8.6 g of a catalyst powder.

Catalyst 4

[0024] To a 5L reactor were fed a mixed solution prepared by dissolving 55.8g of $Zn(NO_3)_2 \cdot 6H_2O$, 112.5g of $Al(NO_3)_3 \cdot 9H_2O$ and 144.2g of $Mg(NO_3)_2 \cdot 6H_2O$ in 1299g of ion-exchange water, a 0.24 mol/L Na_2CO_3 aqueous solution and a 4N NaOH aqueous solution at the rate of 12.5 ml/min, 9 ml/min and 5 to 7.5 ml/min, respectively. The reactor was charged in advance with 500g of water and stirred at 250 rpm by means of a constant speed stirrer. The temperature of the reaction liquid was maintained at $15 \pm 2^\circ C$, and the amount of the NaOH aqueous solution to be added was

controlled to maintain the pH at 9.7 to 10.3 to carry out a precipitation reaction for 2 hours. Then, feeding of the respective aqueous solutions was stopped, and the suspension was aged for one hour with stirring. This suspension was filtered, and the resulting white solid matter was sufficiently washed with ion-exchange water. After washing, the solid matter was dried for 12 hours in a dryer at 110°C to obtain a white solid catalyst precursor represented by the following formula: $[(Zn_{0.25}Mg_{0.75})_{5/7}Al_{2/7}(OH)_2](CO_3)_{1/7} \cdot cH_2O$. Then, the catalyst precursor after drying was calcined at 550°C for 2 hours to obtain a catalyst.

[0025] It is apparent from the researches by W. T. Reichle et al. (Journal of Catalysis, 101, 352-359 (1986)) that the catalyst shown above turns to a complex oxide on these calcining conditions.

10 Example 1

[0026] A 300mL four-necked flask was charged with 3.1 g of catalyst 1 obtained in the catalyst preparation, 35.9 g of glycerin and 60.5 g of 2-ethylhexyl glycidyl ether, and they were reacted at 170°C for 6 hours under a nitrogen atmosphere while stirring to obtain 99.5 g of a mixture containing diglyceryl mono-2-ethylhexyl ether. A result obtained by analyzing the composition of this mixture by means of gas chromatography is shown in Table 1.

15 Examples 2 to 9

[0027] Using Catalysts 2 to 4, a glycidyl ether and an active hydrogen-containing organic compound each in an amount shown in Table 1 were reacted similarly to Example 1 at the temperature and for the time shown in Table 1. The composition and the selectivity of each of the products obtained by analyzing them by means of gas chromatography are shown in Table 1.

20 Comparative Example 1

[0028] A 300mL four-necked flask was charged with 0.4 g of NaOH as a catalyst, 55.3 g of glycerin and 93.1 g of 2-ethylhexyl glycidyl ether, and the reaction materials were reacted at 180°C for 5 hours under a nitrogen atmosphere with stirring to obtain 148.4 g of a mixture containing diglyceryl mono-2-ethylhexyl ether. The composition of this mixture was analyzed by means of gas chromatography, and the result thereof is shown in Table 1.

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Table 1

	Catalyst (g)	Reaction materials (g)		Mole ratio (a)/(b)	Reaction time (hour)	Temperature (°C)
		Glycidyl ether (a)	Active hydrogen-containing organic compound (b)			
Example 1	Catalyst 1 (3.1)	I (60.5)	A (35.9)	0.83	6	170
Example 2	Catalyst 2 (4.7)	I (93.1)	A (55.3)	0.83	6	170
Example 3	Catalyst 3 (3.1)	I (60.5)	A (35.9)	0.83	6	170
Example 4	Catalyst 4 (3.1)	I (60.5)	A (35.9)	0.83	5	170
Example 5	Catalyst 2 (4.7)	I (93.2)	B (124.7)	0.83	9	175
Example 6	Catalyst 2 (2.8)	G (39.1)	A (33.2)	0.83	6	150
Example 7	Catalyst 2 (3.8)	H (75.1)	A (55.3)	0.83	4	180
Example 8	Catalyst 2 (2.8)	J (69.7)	A (33.2)	0.83	20	150
Example 9	Catalyst 2 (3.1)	I (60.5)	C (64.4)	0.67	66	160
Comparative Example 1	NaOH (0.4)	I (93.1)	A (55.3)	0.83	5	180

Table 1 (continued)

	Composition (GC area %)			Selectivity (1)/[(1)+(2)+(3)]
	Active hydrogen-containing organic compound	Monoether product (1)	Diether product (2)	
Example 1	14.9	62.1	17.5	1.4
Example 2	16.1	58.4	20.0	3.6
Example 3	15.9	58.9	19.3	1.8
Example 4	16.7	59.4	19.0	1.8
Example 5	31.1	41.3	12.1	1.8
Example 6	23.6	53.3	16.6	1.9
Example 7	22.0	54.2	19.7	2.4
Example 8	14.9	51.1	17.0	2.3
Example 9	18.7	67.3	1.7	0.3
Comparative Example 1	22.1	43.5	25.3	7.1

A: glycerin
B: diglycerin
C: 2,2-dimethyl-1,3-dioxolane-4-methanol (acetone ketal of glycerin)
G: butyl glycidyl ether
H: phenyl glycidyl ether
I: 2-ethylhexyl glycidyl ether
J: decyl glycidyl ether

5 Examples 10 to 12

10 [0029] Using Catalyst 2, a glycidyl ether and an active hydrogen-containing organic compound each in an amount shown in Table 2 were reacted similarly to Example 1 at the temperature and for the time shown in Table 2. The composition and the selectivity of each of the products obtained by analyzing them by means of gas chromatography are shown in Table 2.

15 Comparative Example 2

20 [0030] NaOH (6.42g) was used as a catalyst, and a glycidyl ether and an active hydrogen-containing organic compound each in an amount shown in Table 2 were reacted in the same manner as in Example 1 at the temperature and for the time shown in Table 2. The composition and the selectivity of the product obtained by analyzing it by means of gas chromatography are shown in Table 2.

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Table 2

	Catalyst (g)	Reaction materials (g)	Active hydrogen-containing organic compound (a) (b)	Mole ratio (a)/(b)	Reaction time (hour)	Temperature (°C)
Example 10	Catalyst 2 (6.42)	K (128.4)	A (27.6)	2.0	6.5	175
Example 11	Catalyst 2 (10.04)	G (36.9)	A (112.2)	2.15	5	160
Example 12	Catalyst 2 (4.65)	I (93.2)	A (23.2)	2.0	6	180
Comparative Example 2	NaOH (0.38)	I (93.2)	A (23.2)	2.0	2	170

Table 2 (continued)

	Composition				Selectivity $(2) / [(1) + (2) + (3) + (4) + (5)]$
	Monoether product (1)	Diether product (2)	Triether product (3)	Tetraether product (4)	
Example 10	4.4	86.6	9.0	0	0
Example 11	15.2	72.6	11.8	0.8	0
Example 12	2.5	76.3	20.8	0.5	0
Comparative Example 2	15.9	41.4	33.4	8.8	0.5
					41.4 %

A: glycerin

G: butyl glycidyl ether

I: 2-ethylhexyl glycidyl ether

K: isodecyl glycidyl ether

Examples 13 to 15

5 [0031] Using Catalyst 2, a glycidyl ether and an active hydrogen-containing organic compound each in an amount shown in Table 3 were reacted similarly to Example 1 at the temperature and for the time shown in Table 3. The composition and the selectivity of each of the products obtained by analyzing them by means of gas chromatography are shown in Table 3.

Comparative Example 3

10 [0032] Using 0.3g of NaOH as a catalyst, a glycidyl ether and an active hydrogen-containing organic compound each in an amount shown in Table 3 were reacted similarly to Example 1 at the temperature and for the time shown in Table 3. The composition and the selectivity of the product obtained by analyzing it by means of gas chromatography are shown in Table 3.

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Table 3

	Catalyst (g)	Reaction materials (g)	Mole ratio (a)/(b)	Reaction time (hour)	Temperature (°C)
	Glycidyl ether (a)	Active hydrogen-containing organic compound (b)			
Example 13	Catalyst 2 (3.0)	I (27.7)	E02 (32.5)	1	5
Example 14	Catalyst 2 (6.0)	I (61.2)	E04 (95.6)	1.06	6
Example 15	Catalyst 2 (3.0)	I (30.4)	E06 (60.5)	1.05	6
Comparative Example 3	NaOH (0.3)	I (27.7)	E02 (32.5)	1	4.5
					110

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Table 3 (continued)

	Composition		Selectivity (1)/[(1)+(2)]
	Monoether product (1)*	Diether product (2)**	
Example 13	97.4	2.6	97.4
Example 14	99.6	0.4	99.6
Example 15	99.9	0.1	99.9
Comparative Example 3	69.5	30.5	69.5

I: 2-ethylhexyl glycidyl ether

E02: polyoxyethylene (n = 2)-2-ethylhexyl ether

E04: polyoxyethylene (n = 4)-2-ethylhexyl ether

E06: polyoxyethylene (n = 6)-2-ethylhexyl ether

*: a compound in which one glycidyl ether (a) is added to compound (b)

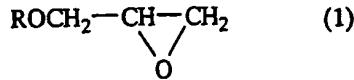
**: a compound in which two glycidyl ethers (a) are added to compound (b)

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[0033] In an addition reaction between an active hydrogen-containing organic compound and a glycidyl ether, use of the preferred catalyst of the present invention enables inhibition of an excess addition reaction of glycidyl ether which is a successive reaction, and a mono- or di(alkyl, alkenyl or phenyl) ether product in which one or two glycidyl ethers are added is selectively obtained by adjusting the mole ratio of the active hydrogen-containing organic compound to the glycidyl ether to fall in a specific range. That is, a mono- or di-(alkyl, alkenyl or phenyl) ether product can be obtained at a high productivity with a high purity.

Claims

1. A catalyst for an addition reaction between an active hydrogen-containing organic compound and a glycidyl ether, comprising a complex oxide of magnesium and at least one element other than magnesium selected from the group consisting of the elements in the third period and the fourth period in the periodic table.
2. The catalyst according to claim 1, comprising a complex oxide of magnesium and at least one element selected from aluminum and zinc.
- 10 3. A process for producing a glycidyl ether adduct, which comprises subjecting an active hydrogen-containing organic compound and a glycidyl ether to an addition reaction in the presence of a catalyst comprising a complex oxide of magnesium and at least one element other than magnesium selected from the group consisting of the elements in the third period and the fourth period in the periodic table.
- 15 4. The process according to claim 3, wherein the active hydrogen-containing organic compound and the glycidyl ether are subjected to the addition reaction in the presence of a catalyst comprising a complex oxide of magnesium and at least one element selected from aluminum and zinc.
- 20 5. The process according to claim 3 or 4, wherein the active hydrogen-containing organic compound is a hydroxyl group-containing compound.
6. The process according to claim 5, wherein the hydroxyl group-containing compound is at least one selected from the group consisting of linear or branched monohydric alcohols having 1 to 30 carbon atoms, polyols having 2 to 18 carbon atoms and 2 to 18 hydroxyl groups and ketals thereof, polyoxyalkylene alkyl ethers, and mixtures thereof.
- 25 7. The process according to any of claims 3 to 6, wherein the active hydrogen-containing organic compound is at least one selected from the group consisting of ethylene glycol, 1,2-propanediol, 1,3-propanediol, glycerin, pentaerythritol, diglycerin, polyglycerin, sorbitol, glucose, sucrose, glycerin ketal, and mixtures thereof.
- 30 8. The process according to any of claims 3 to 7, wherein the glycidyl ether is represented by Formula (1):



wherein R represents a linear or branched alkyl or alkenyl group having 1 to 36 carbon atoms or a phenyl group.

- 40 9. The process according to any of claims 3 to 8, wherein the glycidyl ether adduct is a product in which one glycidyl ether is added.
- 45 10. A process for producing a product in which one glycidyl ether is added, which comprises subjecting a glycidyl ether and an active hydrogen-containing organic compound to an addition reaction in a mole ratio of the glycidyl ether to the organic compound in a range of from 1 : 0.9 to 1 : 3 in the presence of a catalyst comprising a complex oxide of magnesium and at least one element other than magnesium selected from the group consisting of the elements in the third period and the fourth period in the periodic table.
- 50 11. A process for producing a product in which two glycidyl ethers are added, which comprises subjecting a glycidyl ether and an active hydrogen-containing organic compound to an addition reaction in a mole ratio of the glycidyl ether to the organic compound in a range of from 1.8 : 1 to 3 : 1 in the presence of a catalyst comprising a complex oxide of magnesium and at least one element other than magnesium selected from the group consisting of the elements in the third period and the fourth period in the periodic table.



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In the addition reaction between the active hydrogen-containing organic compound and the glycidyl

ether, use of the catalyst of the present invention enables inhibition of an excess addition reaction of the glycidyl ether which is a successive reaction, and a mono- or di-(alkyl, alkenyl or phenyl) ether product in which one or two glycidyl ethers are added is selectively produced by adjusting the mole ratio of the active hydrogen-containing organic compound to the glycidyl ether to a specific range. Accordingly, the mono- or di-(alkyl, alkenyl or phenyl) ether product can be obtained at a high productivity with a high purity.



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PARTIAL EUROPEAN SEARCH REPORT

Application Number

which under Rule 45 of the European Patent Convention EP 03 01 9511
shall be considered, for the purposes of subsequent
proceedings, as the European search report

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
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X	US 5 686 379 A (IMANAKA TATSUHIRO ET AL) 11 November 1997 (1997-11-11) * column 2, line 60 - column 3, line 2; examples 1,3,6; table 1 * * column 6, line 30 - column 7, line 34 * * column 3, line 19 - line 23; claims 10,11 * -----	1,2	B01J21/00 B01J21/10 C11D1/72
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X,D	REICHLE, W.T. ET AL.: "The nature of the thermal decomposition of a catalytically active anionic clay mineral" JOURNAL OF CATALYSIS, vol. 101, 1986, pages 352-359, XP009036728 * abstract * ----- -----	1,2	
		-/-	TECHNICAL FIELDS SEARCHED (Int.Cl.7) B01J C11D C08G C07C
INCOMPLETE SEARCH			
<p>The Search Division considers that the present application, or one or more of its claims, does/do not comply with the EPC to such an extent that a meaningful search into the state of the art cannot be carried out, or can only be carried out partially, for these claims.</p> <p>Claims searched completely :</p> <p>Claims searched incompletely :</p> <p>Claims not searched :</p> <p>Reason for the limitation of the search:</p> <p>see sheet C</p>			
Place of search	Date of completion of the search		Examiner
Munich	22 September 2004		Bork, A-M
CATEGORY OF CITED DOCUMENTS			
<p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p>		<p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>	

European Patent
OfficeINCOMPLETE SEARCH
SHEET C

Application Number

EP 03 01 9511

Claim(s) searched completely:
2,4

Claim(s) searched incompletely:
1,3,5-11

Reason for the limitation of the search:

Present claim 1 relate to an extremely large number of possible catalysts for use in a process for producing a glycidyl ether adduct. Support within the meaning of Article 84 EPC and/or disclosure within the meaning of Article 83 EPC is to be found, however, for only a very small proportion of the catalysts claimed.

Based on the world-wide accepted postulate that the catalytic properties of a compound are considered to be unexpected by the skilled person, the broad definition of the catalyst defined in claim 1 is considered as highly

speculative and not supported by the description.

Consequently, the search has been carried out for those parts of the claims which appear to be supported and disclosed and for which a catalytic effect was proven, namely those parts relating to catalysts comprising a complex oxide of magnesium and at least one element selected from aluminium and zinc, according to claim 2.

For the same reasons the processes according to claims 3, 10 and 11 were restricted to a process using catalysts comprising a complex oxide of magnesium and at least one element selected from aluminium and zinc, according to claim 4.



European Patent
Office

PARTIAL EUROPEAN SEARCH REPORT

Application Number

EP 03 01 9511

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
			TECHNICAL FIELDS SEARCHED (Int.Cl.7)
X	PATENT ABSTRACTS OF JAPAN vol. 015, no. 181 (C-0830), 9 May 1991 (1991-05-09) & JP 03 044354 A (DAICEL CHEM IND LTD), 26 February 1991 (1991-02-26) * abstract * -----	1,2	
X	PATENT ABSTRACTS OF JAPAN vol. 011, no. 163 (C-424), 26 May 1987 (1987-05-26) & JP 61 291549 A (NIPPON NOHYAKU CO LTD), 22 December 1986 (1986-12-22) * abstract * -----	1,2	
X	US 4 970 191 A (SCHUTZ ALAIN A) 13 November 1990 (1990-11-13) * column 2, line 3 - line 20 * * column 4, line 21 - line 24; claims 1,4 *	1,2	
A	US 4 465 866 A (URATA KOUICHI ET AL) 14 August 1984 (1984-08-14) * the whole document *	1-11	
A,D	WO 93/09214 A (PROCTER & GAMBLE) 13 May 1993 (1993-05-13) * page 1 - page 5; examples 1,2 *	1-11	

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EP 03 01 9511

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22-09-2004

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